The Study of Chain Initiation Effect on the Direct Initiation of Detonation

Masoud Afrand, Saeid Farahat, and Mehdi Alamkar

Abstract—In this research, effect of combustion reaction mechanism on direct initiation of detonation has been studied numerically. For this purpose, reaction mechanism has been simulated by using a three-step chemical kinetics model. The reaction scheme consists sequentially of a chain-initiation and chain-branching step, followed by a temperature -independent chain-termination. In a previous research, the effect of chain-branching on the direct initiation of detonation is studied. In this research effect of chain-initiation on direct initiation of detonation is investigated. For the investigation, first a characteristic time (τ) for each step of mechanism, which includes effect of different kinetics parameters, is defined. Then the effect of characteristic time of chain-initiation (τ_I) on critical initiation energy is studied. It is seen that increasing τ_I, causes critical initiation energy to be increased. Drawing detonation's shock pressure diagrams for different cases, shows that in small value of τ_I, kinetics has more important effect on the behavior of the wave.

Keywords—Detonation initiation, Initiation energy, Reaction rate, Characteristic time.

I. INTRODUCTION

IN general, a combustible mixture can support two modes of combustion: deflagration and detonation. In the deflagration regime, in the limit it can burn a s a laminar flame at a typical velocity of the order of about 0.5 m/s, or it may accelerate to a turbulent flame where velocity can be orders of magnitude higher. The other extreme is the detonation mode, in which a detonation wave propagates at about 2000 m/s amplifying the pressure by a factor of 20 across the wave. It is interesting to note that a diverse variety of propagation mechanisms are responsible for the observed four orders of magnitude change in wave velocity. While the propagation of a laminar deflagration is governed by the molecular diffusion of heat and mass from the reaction zone to the unburned mixture, the propagation of a detonation depends on the adiabatic shock compression of the unburned mixture. To support a detonation at such high speed, the rate of transformation of energy in the reaction zone must be very high. The combustible gas immediately ahead of the reaction front must be heated to such a high temperature as to permit a high reaction rate in the reaction front. This thesis deals with the second mode, i.e.

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gaseous detonation waves. Detonation is defined as a combustion-supported shock wave with significant pressure and density rise across the wave. The main characteristic of detonation waves is the coupled motion of the shock front and the reaction zone behind it.

Generally, there are two modes of initiation. The combustible mixture can be ignited by a low energy source and burn as a slow flame. Under appropriate conditions, it will accelerate and undergo a transition to detonation. This process is referred to as deflagration to detonation transition (DDT). During DDT the mechanism of the propagation of combustion wave changes from molecular diffusion to convective turbulent transport and auto ignition by adiabatic shock compression. In the final phase of DDT, the shock-flame complex is observed to propagate at about half the CJ detonation velocity for a certain period of time, which terminates with the abrupt onset of a detonation wave. This regime of propagation is called the quasi-steady regime. The ignition source plays no role in the DDT process. Comprehensive reviews on DDT have been published by Lee et al. [1980], Shepherd et al. [1992], and Sichel [1992]. [5]

The second mode of initiation, direct initiation, is also referred to as the fast mode of generating a detonation wave, where the detonation is formed instantaneously via the rapid deposition of a large amount of energy in a small volume of the combustible mixture. If a sufficient amount of energy is released by the igniter, rapid auto ignition takes place behind the generated blast wave and the reaction-coupled shock quickly becomes a CJ detonation. For direct or blast initiation, the energy of the source is the sole parameter that governs success or failure of detonation initiation. When the critical initiation energy is deposited in a mixture, initially a highly overdriven detonation is formed, where the shock front and the reaction front (the flame) are coupled and move together. Then, the wave decays to a strength about half the CJ velocity in which the shock and the reaction front are decoupled. For a period of time, much longer than the chemical induction time, the shock propagates almost steadily. This so-called quasi-steady period is terminated with an abrupt acceleration of the shock front to an overdriven detonation wave. Finally, the overdriven wave decays to a self-sustained detonation wave. If the initiation energy is slightly smaller than this critical value, the decoupling continues and the shock eventually decays to an acoustic wave.

The first work to predict the critical initiation energy was the pioneering study of Zeldovich *et al.* [1956]. They stated that for successful initiation, the shock should have a minimum strength (Le., CJ strength) when it has propagated a

distance at least of the order of the chemical induction length. They proposed a criterion which is still the framework of many current theories. This criterion shows the dependence of the critical initiation energy, E_C, to chemical induction length, A, (i.e., $E_C \propto \Delta^3$ for spherical detonations). Since in this criterion the constant of proportionality is missing, it has been the subject of further research. [5]

To survey the detonation phenomenon a lot of studies have used the simple one-step model with Arrhenius reaction rate to model the combustion process. The investigations of Mazaheri [1], Sharpe [2] and Eckett[3] are included. Because of its application simplicity the one-step model is very suitable but it has some inconveniencies. For example this model is not capable to model induction length independently and detonation decay could not be also modeled. Short and Quirk[4] have used a three-step model (included initiation, branching and termination steps) to study stability of detonation. This three-step model which is similar to actual mechanism has not the inconveniences of one-step model. Since in a reaction mechanism chain branching is the most important one, Short and Quirk have studied the effect of chain branching on stability of detonation.

Showing the role of chain initiation, Mazaheri and Hashemi [5] have investigated the role of this chain on the stability of detonation. Lee and NG [6] studied the role of chain branching on direct initiation of detonation by making use of three-step model. In this survey they have observed the different regimes of critical, sub-critical and super-critical by the change in chain branching rate. They concluded that the three-step mechanism presents a more precious result in comparison with one-step mechanism.

Regarding that in the previous work, the chain initiation has not been studied, in this study the role of this parameter on the direct initiation of detonation is investigated.
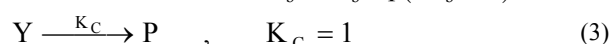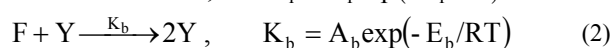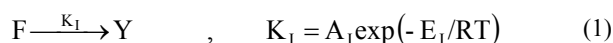
II. CHEMISTRY MODEL

In a numerical simulation of gaseous detonations, the closure of the conservation equations is attained by the prescription of an equation of state and a reaction model. The issue of the reaction model is addressed here.

Choices for the reaction model include a vast range from a single step model to a detailed chemical-kinetics mechanism. Due to the simplicity and computational efficiency of the one-step model, this model has been used extensively in previous research. Although this simple model is able to capture many generic features of gaseous detonations, it has several drawbacks. For example, it contains no induction length, which is known to control much of the dynamic behavior of a detonation [3].

This model provides no mechanism for the quenching of reactions, and causes any initiation simulation, eventually, to produce a detonation [1]. On the other hand, the detailed reaction mechanism is computationally very expensive. To overcome this dilemma, several reduced as well as global mechanisms have been proposed in previous research. In reality, chemical kinetics consists of a sequence of chain-initiation, chain-propagation, chain-branching, and chain-termination stages. In such mechanisms, a small amount of the reactant mixture is converted into chain carriers (i.e., radicals) by means of relatively slow and high-activation energy initiation process. The radicals are then rapidly multiplied through a sequence of self-accelerating chain-branching reactions. At the same time, the rise of the concentration of radicals is retarded by termination reactions. Based on a chain-branching model of Gray and Yang (1965) and its later generalization, a three-step model was employed by Dold and Kapila (1991). Comparing the one-step Arrhenius kinetic with the three-step model, they showed that a quite different route to detonation initiation exists. This model has been used later in many investigations (e.g., Short and Quirk, 1997). The chemical kinetic model used for present study is a generalized three-step chain-branching reaction model and its detailed description can be found in the paper by Mazaheri & Hashemi[5]. Which contains chain initiation, chain branching, and chain termination steps, as follows:

$$F \xrightarrow{\ K_I\ } Y \qquad , \qquad K_I = A_I \exp(-E_I/RT) \qquad (1)$$

$$F + Y \xrightarrow{\ K_b\ } 2Y , \qquad K_b = A_b \exp(-E_b/RT) \qquad (2)$$

$$Y \xrightarrow{\ K_C\ } P \qquad , \qquad K_C = 1 \qquad (3)$$

where F, Y, and P represent the reactant, radical, and product of reaction.

K_I is the rate constant of each reaction, and, A and E are the constants of the Arrhenius model. There is a simple relation between the crossover temperature (T_{cr}) of Short and Quirk (1997) and the parameter A_b in present analysis; that is: $A_b = \exp(E_b/T_{cr})$.[5]

The rates of the above reactions are $r_I = k_I f$, $r_b = \rho K_b f y$, and $r_c = K_c y$, where f and y are the mass fraction of fuel and radical, respectively. In order to study the effect of the initiation step on the behavior of detonation, an individual time scale is defined for each reaction step. These characteristic times are defined as:

$$\tilde{\tau}_I \propto \frac{1}{r_I} \approx \frac{1}{A_I \exp(-E_I/RT_s)} \qquad (4)$$

$$\tilde{\tau}_b \propto \frac{1}{r_b} \approx \frac{1}{\rho_s A_b \exp(-E_b/RT_s)} \qquad (5)$$

$$\tilde{\tau}_C \propto \frac{1}{r_C} \approx \frac{1}{A_C} \qquad (6)$$

Subscript ''s'' in the above relations denotes the post shock condition. These characteristic times are used as the main kinetic parameters in this study.

III. CONSERVATION EQUATIONS

Ignoring viscosity, conductive heat transfer, diffusion, and body forces, the governing equations for compressible reactive flow are reactive Euler equations. If the multidimensional

character of a detonation can be also ignored, then a one-dimensional description is valid. In a fixed reference frame, the one-dimensional reactive Euler equations for a planar geometry are given by:

$$\frac{\partial \rho}{\partial t} + \frac{\partial(\rho u)}{\partial x} = 0$$

$$\frac{\partial(\rho u)}{\partial t} + \frac{\partial}{\partial x}(\rho u^2 + p) = 0$$

$$\frac{\partial(\rho u)}{\partial t} + \frac{\partial}{\partial x}[u(\rho e + p)] = 0 \qquad (7)$$

$$\frac{\partial(\rho f)}{\partial t} + \frac{\partial(\rho uf)}{\partial x} + = \rho w_F$$

$$\frac{\partial(\rho z)}{\partial t} + \frac{\partial(\rho uz)}{\partial x} + = \rho w_P$$

where variables ρ, u, p, and e are the density, particle velocity, pressure, and total energy, respectively. w_F is the consumption rate of the reactant, and wP is the production rate of the product. The mass fraction of the radicals is obtained from $y = 1 - f - z$. A callorically perfect gas equation of state is used throughout the study. Therefore:

$$e = \frac{p}{\rho(\gamma - 1)} + \frac{u^2}{2} - Q(1 - z), \quad P = \rho RT \qquad (8)$$

where Q is the heat release per unit mass of the reactant and c is the ratio of the specific heats. The dependent variables are non-dimensionalized with respect to the unburnt mixture properties. Thus, the density, pressure and velocity are non-dimensionalized with ρ_0, γP_0 and the sound speed of unburnt mixture (C_0), respectively. The termination step characteristic time ($\tilde{\tau}_C$) is chosen as the time scale t_c for non-dimensionalization. Therefore, the non-dimensional termination characteristic time becomes unity .To non-dimensionalize distances, the reference length (L_c) is setto tc times the sound speed of the unburnt mixture. Q and Ea are non-dimensionalized with RT_0.

IV. NUMERICAL METHOD

Over the past 40 years, a great number of numerical schemes have been devised for the simulation of high speed compressible flows with strong shock waves. In recent years, a number of new shock-capturing schemes, often called high-resolution schemes, have been proposed. Among them are the FCT, MUSCL, ENO, and PPM methods. There are several excellent review articles which compare these schemes from different point of views. Interested readers are referred to those articles, particularly the paper of Yang et al., and the Ph.D. thesis of Bourlioux[7]. After comparing different schemes, they recommended the PPM (Piecewise Parabolic Method) of Colella and Woodward [8] as the best in overall performance. Therefore, in the present work, PPM is chosen as the main gasdynamics solver. PPM is a third order method near the discontinuities and forth orders in smooth regions [8]. The developed code is validated via several test problems [1].

It is controlled that the results are not dependent on the grid in all calculation series and the optimum grid is used.

V. RESULTS AND DISCUSSION

The initial blast wave is modeled as the early situation through the model of the ideal and strong explosive wave which Mazaheri and Eckett have applied this method. For ideal strong blast waves, the initial conditions are given by the similarity solution of Taylor [5] and Sedov[5]. For a perfect gas with constant specific heat ratio γ, the similarity solution of the point blast model consistent with earlier normalization is given by:

$$u_s = \frac{2}{\gamma+1}M_s \quad , \quad p_s = \frac{2}{\gamma+1}M_s^2 \quad , \quad \rho_s = \frac{\gamma+1}{\gamma-1}$$

$$\frac{u}{u_s} = f\left(\frac{r}{R_s}\right) \quad , \quad \frac{\rho}{\rho_s} = g\left(\frac{r}{R_s}\right) \quad , \quad \frac{p}{p_s} = h\left(\frac{r}{R_s}\right) \qquad (9)$$

where subscript s refers to the conditions immediately after the shock. M_s and R_s respectively, are defined as the shock mach number shock radius where the place distribution of variable of back ward conduct of the blast wave (including , velocity u. density ρ. pressure p), can be obtained by solving the functions $f(r/R_s)$, $g(r/R_s)$ and $h(r/R_s)$, which are listed in the book by Korobeinikov (1991) [11]. In addition, the similar solution about the potent irruptive (blast) wave has been handed out a respect in which shock (M_s) power and shock (R_s) radius as well as the dimensionless initiation energy in succession, has influenced on also area unit and length unit which are shown in the three plate, cylindrical and spherical geometries:

$$E_s = \alpha_j \left(\frac{j+3}{2}\right)^2 \gamma M_s^2 R_s^{j+1} \qquad (10)$$

In this regard, α_j is considered energy invariable integral. In plate geometry α= 2.557 which has been figured out by korbeinikov in 1991[11]. Moreover, $\tilde{p}_0$ is the initial mixed pressure (dimension able) and j is confined to zero for plate, one in cylindrical geometrical sheet, and two in spherical geometry. Knowing M_s and R_s, initial energy can be determined according to relation .9.

The selective variables of kinetic parameters are included (Ac=1, Ap=20 $A_1=10^3$, $A_b=16000$, $E_b=15$, $E_p=8$). the activation energy of the chain initiation is varied, since the chain initiation characteristic time to be changed. The dimensionless variables of the characteristic times are achieved through the exertion of variables which are fixed and consistent during the calculation. The answer is accomplished in the below simplified relation:

$$\tau_I = \frac{\exp(E_I/1.89)}{1000} \quad \text{‹} \tau_b = 0.033 \quad \text{‹} \tau_C = 1$$

It is essential to note that during the investigation of Ng, lee the effect of variation of τ_b was studied. The more τ_b rises, the

detonation more likely tend to proceed instability (at this state, it leads to fluctuation behavior) and eventually, it approaches to decay situation. According to our purpose in this essay which aims to figure out the effect of τ_I variation, a little variable was chosen for τ_b to provide a wider range for the variations of τ_I variable. The rise of τ_I variables continues as long as detonation is not established in the mixture any more i.e. we reach to a so-called detonability limit of the mixture. This survey is accomplished in several parts. In the first part we concern to investigation of effect of the rise of initiation energy and observation of critical ignition situation. In the second part we study the variation of amount of critical initiation energy as an effect of variation of τ_I. Finally we will study the effect of changes in τ_I on the attitude of detonation wave.

A. The Investigation of the Effect of Initiation Energy Rise on the Direct Initiation with $\tau_I = 6.13 \times 10^{10}$

In this section, the investigation of the initiation energy rise effect within above–mentioned τ_I is more underscored. Initially, the energy is assumed in a way that detonation proceeds decay condition, then, we approach a boundary at which detonation is formed through the gradual increase of energy. The initiation energy at this boundary is the same as critical and initiation energy.

Since the figure 1 shows that detonation in $E_0 = 1141.84$ tends to decay. It means that the amount of initiation energy is less than the limit which would form the detonation. This Condition is called subcritical condition. While the initial energy increases to $E_0 = 1142.14$it is observed that in spite of intensive fluctuations and the tendency of detonation toward decay finally detonation will formed. In fact, this is the limit of critical initiation energy. By the growth of the initiation energy to $E_0 = 1142.44$, it can be noticed that the total behavior of detonation is like the one in previous case, with a distinction, that is the maximum of generated leap is slightly more than the leap took place in the previous case, and the stable behavior is also achieved provided slightly sooner. Continuing the rise in the initiation energy, the tendency of the detonation toward decay will be faded and decreased as well.

The reason of decay of detonation in a case that the initiation energy is assumed lower than the critical limit, is that the rate of reaction due to the descant of shock pressure, is lowered to a level at which the energy, made by reaction, is unable to amplify the shock and eventually, decay procedure is triggered where no pressurized wave is transmitted from the reaction toward shock and therefore, the shock pressure loss goes ahead. The reaction rate decreases according to both shock pressure loss and shock temperature loss and therefore, the distance between reaction front and the shock widely expands, and this separation results in decay detonation. The reason of the establishment of detonation, in a case that the initiation energy tends to rise, is that, the larger initiation energy causes a larger shock pressure during the imitation step. If the combustion reaction strengthens the shock pressure, the temperature and pressure, located behind shock, should not be lower than a limit. When the detonation goes to

decay, the pressure and the temperature behind the shock are lower than this limit.

These results can be taken into consideration from various angles, Lee and NG presented these outcomes in the studying procedure of the initiation of detonation:

"To achieve a successful initiation, the temperature behind shock in the blast wave of initial source, should not be lower than cross over temperature."

Cross over temperature is a temperature at which chain branching rate preserves equality with the chain termination rate. In fact, at this temperature, the rate of both radical production and radical consumption are equal and this matter prevents from the chain blast took place by chain branching and the rate of combustion reaction intensively decreases.

The dimensionless cross over temperature, for this part, is obtained 1.195, which corresponds to a shock pressure equal to 4.2. According to the figure 1 it can be noticed that shock pressure in the blast wave for a case in which the initiation doesn't happen is about 6.5 which is higher than the mentioned number (4.2), thus the mentioned criterion (4.2), at least, will not be valid during all characteristic times.
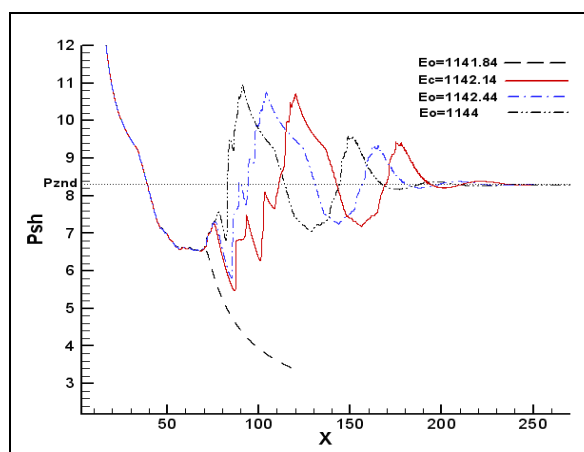

Fig. 1 Effect of initiation energy on shock pressure,
$$\tau_I = 6.13 \times 10^{10} , \ \tau_B = 0.033 , \ Q = 10.0 , \ \gamma = 1.2$$

In figure 2, the effect of the initiation energy rise on shock pressure versus distance in the direct initiation for initiation energies which are higher than critical limit has been investigated. It can be seen in this figure that the more the initiation energy increases, minimum produced pressure in shock wave will be increased. Moreover, the existed leaps will take place in the distance where is farther from center point.

The reason of this phenomenon is that the progressive agent of shock wave before achieving the initial leap is initiation energy, and the larger the amount of this energy, the farther the initial leap will occur and shock ware will move a greater distance with the support of the initiation energy. It is essential to mention that kinetics role appears after the initial pressure loss, namely, after the pressure descended down the P_{znd}, it is the reaction kinetic which is the agent of pressure rise and detonation drive, therefore, if the initial pressure loss be as large as that reduces the reaction rate to a level in which combustion energy is not able to strengthen shock, detonation will decay.
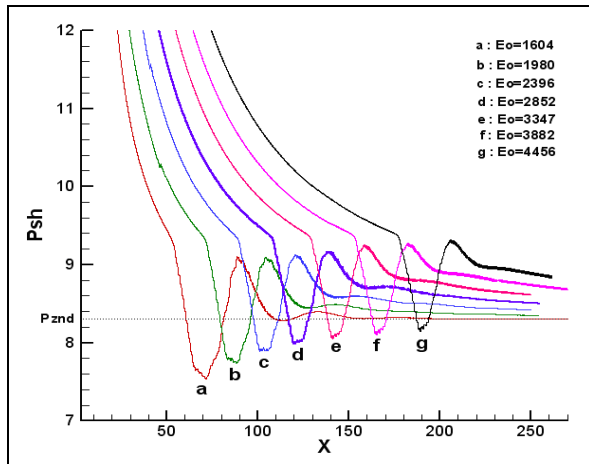
Fig. 2 Effect of initiation energy on shock pressure,
$\tau_I = 6.13 \times 10^{10}$, $\tau_B = 0.033$, $Q = 10.0$, $\gamma = 1.2$

B. The Investigation of the Effect of Initiation Energy Rise on the Direct Initiation with $\tau_I = 8.63 \times 10^{11}$

In this section, the amount of characteristic time of chain initiation is increased from $\tau_I = 6.13 \times 10^{10}$ to $\tau_I = 8.63 \times 10^{11}$. The limit of critical initiation of detonation is shown in figure 3 for this case. As seen, detonation in $E_0 = 2119.5$ tends to decay. With the rise of the amount of the initiation energy to $E_0 = 2120.35$, it can be seen that detonation will be formed which this amount is the same as the critical initiation energy. In contrary to figure 1, fluctuations in this case, has lower intense, in addition, the amplitude of fluctuations are less than amounts that are seen in the figure 1.

In the previous section, as noticed, for $\tau_I = 6.13 \times 10^{10}$ (figure 1) shock pressure loss during the initial step continues to $P_{min} = 6.6$. In contrary to figure 1, in figure 3 in $\tau_I = 8.63 \times 10^{11}$ the pressure loss goes ahead to $P_{min} = 7.6$. According to the figure 3 for $\tau_I = 8.63 \times 10^{11}$, if shock pressure were less than 7.6 ($P_{sh} < 7.6$), the detonation will not be certainly established.

However, as seen in figure 1 in $\tau_I = 6.13 \times 10^{10}$ shock pressure was less than 7.6 ($6.6 < P_{sh} < 7.6$). Detonation was finally organized. Due to the different characteristic times of chain initiation in both two cases, the reason of this phenomenon refers back to the role of the chain initiation too. The reason of this phenomenon is that the chain initiation rate in the case of ($\tau_I = 6.13 \times 10^{10}$), is more than the chain initiation rate in the case of ($\tau_I = 8.63 \times 10^{11}$). The reaction, bearing higher chain initiation rate, could establish required pressure pulse, by lower shock power, for strengthening detonation. In fact, in either case, there is a minimum of temperature which is capable of activating the chain initiation rate adequately for the pressure pulse establishment.

In figure 4, the effect of the initiation energy rise on shock pressure versus of distance in the direct initiation for initiation energies which are very higher than critical limit, has been

investigated. As it is seen in the figure, the existed leaps take place at a distance that is farther than the center point.

In order to survey the effect of τ_I change in constant E_0, a comparison can be made between the figures of 2 and 4. For a better comparison, figure 5 is used which is drawn for $E_0 = 2396$.

Curve c depicts the shock wave behavior in $\tau_I = 6.13 \times 10^{10}$, also curve b displays the shock wave behavior t in $\tau_I = 8.63 \times 10^{11}$. As seen, P_{min} equals 7.9 in the curve C, while p_{min} has the amount of 7.7 in the curve b, thus as a conclusion, in a constant E_0, the amount p_{min} reduces in the in initial pressure loss of initiation by the rise of τ_I. In a case, in which τ_I is smaller, the chain initiation rate will be higher and the required pressure pulse for strengthening of shock wave will be sooner provided[1].

As mentioned previous section, one of the characteristic of critical initiation is that the initial pressure loss of shock front was less than ZND pressure, and then it rose again. According to the conclusion gained in this section, the initial pressure loss decreases through the rise of τ_I. In the limit case for higher τ_I the critical initiation takes place without the initial pressure loss in the blast wave which has not been noticed in alternative investigations.
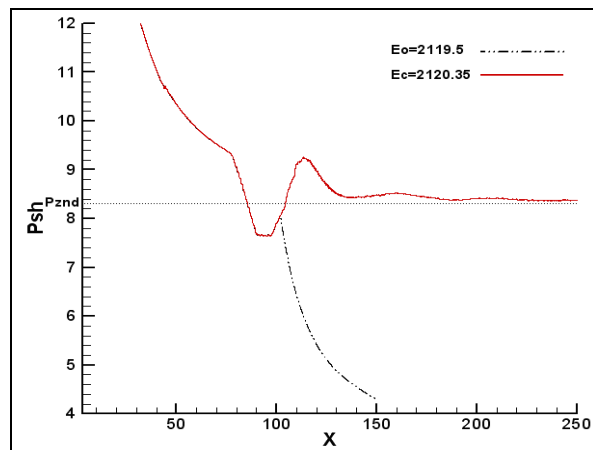

Fig. 3 Effect of initiation energy on shock pressure and critical initiation energy,
$\tau_I = 8.63 \times 10^{11}$, $\tau_B = 0.033$, $Q = 10.0$, $\gamma = 1.2$

C. The Procedure of the Critical Initiation Energy Variation by Characteristic Time of Chain-Initiation (τ_I)

The critical initiation energy amounts which have been obtained for the characteristic time of chain initiation are shown in figure 6. In this diagram the horizontal axis delineating the characteristic time of chain initiation which has been divided logarithmically. The vertical axis depicts the amount of the critical initiation energy. It is noticed that the critical initiation energy increases through the rise of the characteristic time of chain initiation. The increase has been intensified in the higher characteristic time of chain initiation

[1]- *The expression of reason, in more details, requires deeply more study of the detonation structure which is out of our discussion.*

and eventually detonation is not basically established in the mixture for τ_I s bigger than 5×10^{12} (the mixture detonability limit).
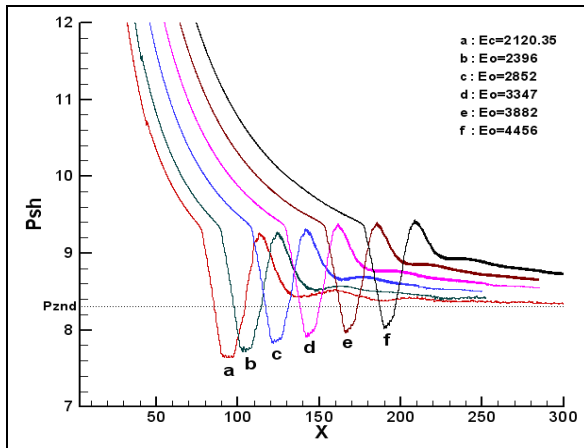

Fig. 4 Effect of initiation energy on shock pressure,
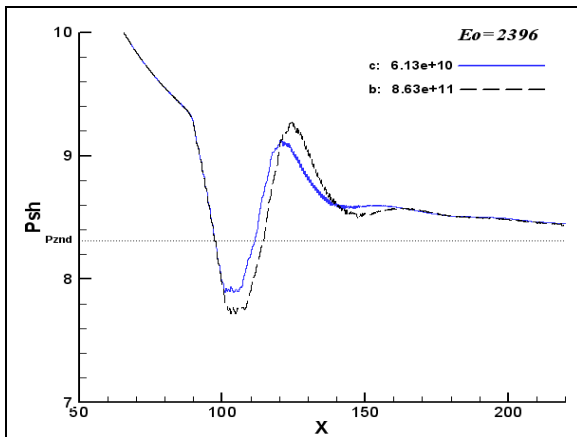$\tau_I = 8.63 \times 10^{11}$, $\tau_B = 0.033$, $Q = 10.0$, $\gamma = 1.2$

Fig. 5 Effect of characteristic time of chain-initiation (τ_I) on P_{min},

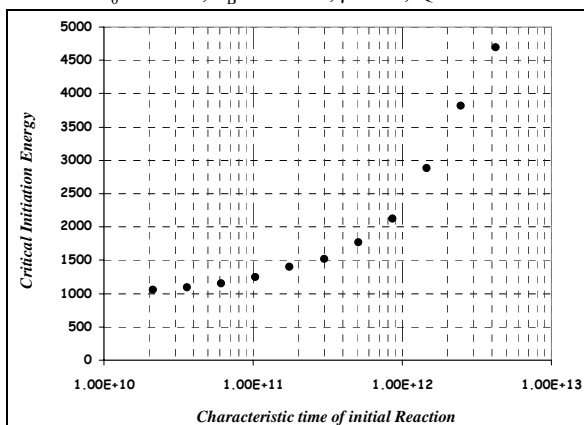$E_0 = 2396$, $\tau_B = 0.033$, $\gamma = 1.2$, $Q = 10.0$

Fig. 6 Effect of characteristic time of chain-initiation (τ_I) on critical initiation energy

$\tau_B = 0.033$, $Q = 10.0$, $\gamma = 1.2$

VI. CONCLUSION

The most significant results gained through the survey of the initiation energy change effect and the characteristic time of the chain initiation on the direct initiation of detonation for $\tau_b = 0.033$ is including:

- The critical initiation energy rises through the characteristic time of chain initiation (τ_I).

- In a constant E_0, the amount of P_{min} is fallen in the initial pressure loss procedure by the rise of τ_I. In a case where τ_I is smaller, the chain initiation rate is higher and the required pressure pulse, for strengthening of shock wave, will be organized sooner. The discrepancy between P_{min}'s for two different characteristic times is smaller. the dependence of minimum shock pressure on the characteristic time of chain initiation is reduced through the initiation energy rise There for, relying of the minimum of shock pressure upon the stated time of the early chain is come down through the rise of the initial energy. In fact, with E_0 rise, shock strength in the initiation step, becomes more than required minimum for the activation of the chain initiation. When the strength trespasses from the minimum level, the role the chain initiation is decreased.

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